

RESPONSE UNDER 37 C.F.R. 1.114(d)  
U.S. Application 09/305,019

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### **REMARKS**

Reconsideration and withdrawal of the rejections of record are respectfully requested.

#### ***Summary of Status of Amendments and Office Action***

Claim 4 to 10 are presently pending in the application, with claim 4 being independent. No amendments to the claims are being made.

The Examiner has rejected claims 4 to 9 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and has rejected claim 10 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and further in view of Innes et al., U.S. Patent No. 4,891,458.

#### ***Response to the Rejection of Claims 4 to 9***

Claims 4 to 9 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Cheng et al., U.S. Patent No. 5,557,024, in view of Kuchenmeister et al., EP 733,608.

In making this rejection, the Examiner stated with respect to independent claim 4:

Cheng discloses alkylation and transalkylation processes in which a feed containing an aromatic compound (e.g., benzene) and olefins (e.g., ethylene) is contacted with MCM-56 or beta zeolite catalyst in an alkylation zone to produce monoalkylbenzene products such as ethylbenzene. By-products (e.g., polyalkylbenzene), which are produced in the alkylation step, can be recycled to the alkylation reactor to undergo transalkylation or they can be reacted with additional aromatic feed in a separate reactor in the presence of TEA-mordenite catalyst to produce monoalkylbenzene products. Cheng discloses reaction conditions for the alkylation step wherein the reaction takes place under liquid phase conditions at temperatures between 150 and 260°C and pressures up to 3000 psig in a space velocity of from 0.01 to 20 WHSV, based on ethylene feed. Cheng also indicates that the ratios of benzene to ethylene in the alkylation reactor may be from 1:1 to 30:1 molar. Cheng further discloses that the transalkylation zone is operated in a stoichiometric excess of aromatic compounds. Therefore, the ratio of benzene to polyalkylbenzene is greater than one. (See col. 3, lines

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25-27; col. 5, lines 19-25; col. 6, lines 1-3; col. 12, line 10 through col. 14, line 40).

The Examiner also rejected dependent claims 5 to 9 over the same combination of references. For purposes of response here, Applicants will direct attention to independent claim 4.

Applicants respectfully submit that there is no *prima facie* case of obviousness, i.e., there is no suggestion to combine the teachings of Cheng et al. with those of Kuchenmeister et al. Further, if a *prima facie* case of obviousness has been established, which it has not, then Applicants have rebutted that case with evidence of unexpected results.

Cheng et al, column 13, line 40 to column 14, line 44, teach a two step alkylation process utilizing two stages, wherein an aromatic feed (benzene) and olefin are reacted in the presence of an alkylation catalyst (such as MCM-56) to produce polyalkylated product and then reacting the polyalkylated product and additional aromatic feed (benzene) in the presence of "a suitable transalkylation catalyst" (column 14, line 27). "The transalkylation catalyst may be a catalyst comprising a zeolite such as MCM-49, MCM-22, PSH-3, SSZ-25, zeolite X, zeolite Y, zeolite beta, or mordenite," (emphasis added) including a TEA-mordenite, Cheng et al. column 14, lines 27-39. Cheng et al., however, do not set forth the particle size of their transalkylation catalyst, nor do they indicate any preference for the use of a TEA-mordenite. Rather, at best Cheng et al. indicate that any transalkylation catalyst can be utilized. Referring to the claims for guidance with respect to preferences, as would be common for one of ordinary skill in the art, it is noted that dependant claim 3 lists MCM-22, zeolite X, zeolite Y, zeolite beta, and mordenite; dependant claims 6 and 9 require zeolite beta.

Kuchenmeister et al, page 2, lines 12 to 28, described the problem being solved as excessive production of polyalkylbenzenes at high temperatures and pressures associated with vapor phase reactions. Kuchenmeister et al, page 2, lines 19 to 44, and the conditions disclosed on page 3, lines 20 and 43 to 44, teach a method for transalkylating polyalkylbenzenes in the vapor phase with a catalyst which has a sodium content of less

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than 50 ppm. At page 3, lines 19 to 22 and lines 29 to 35, Kuchenmeister et al. teach that "the catalyst" used in this invention is selective to the production of ethylbenzene at conditions of about 250 to about 500°C at pressures from about 200 to about 500 psi, is primarily monoclinic aluminosilicate, but can contain up to about 40% orthorhombic crystalline structure, and has a crystal size of less than about 0.5 micron.

Applicants submit that there is no *prima facie* case of obviousness because there is no motivation to combine the teachings of Cheng et al. with those of Kuchenmeister et al. If a *prima facie* case of obviousness has been established, which it has not, then Applicants have rebutted that case with unexpected results.

Applicants submit that there is no suggestion to modify Cheng et al. with Kuchenmeister et al. Cheng et al. broadly teach that any transalkylation catalyst including TEA-mordenite may be used as a transalkylation catalyst in the Cheng et al. process. Kuchenmeister et al. teach that their transalkylation catalyst is primarily monoclinic aluminosilicate, but can contain up to about 40% orthorhombic crystalline structure. Applicants submit that Kuchenmeister et al. teach away from the use of a mordenite catalyst, since mordenite has an orthorhombic crystalline structure and Kuchenmeister et al. place an upper limit of 40% on the percentage of orthorhombic structure which can be contained in the catalyst of their invention. In this regard, the Examiner's attention is directed to Meir and Olson, "Atlas of Zeolite Structure Types," Appendix A, page 144 (Butterworth-Heinemann 1992).

Kuchenmeister et al. further place a limit on the silica to alumina ratio, specifying a Si/Al atomic ratio of 50 to 500. The Examiner's attention is directed to Table 1 of the attached Declaration by Dr. Wieslaw J. Roth under 37 CFR 1.132 which indicates that the TEA-mordenite of the present invention has a Si/Al ratio of 34. This Si/Al ratio outside the range of the teachings of Kuchenmeister et al. further teaches away from the use of TEA-mordenite.

If the teachings of Kuchenmeister et al. were incorporated into Cheng et al., Kuchenmeister et al. would teach a transalkylation process in the vapor phase using a catalyst having a sodium content less than 50 ppm, a monoclinic structure, and a Si/Al molar ratio of 50 to 500. Applicants submit that the mordenite described in Cheng et al.

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has not been shown to have these properties. It is Applicants' position that the mordenite of Cheng et al. would not have the aforementioned properties. See Meir and Olson publication, *supra*. Further, if Kuchenmeister et al. were combined with Cheng et al., the combination would result in a transalkylation process being conducted in the vapor phase in contrast to the claimed invention which recites that transalkylation is conducted in the liquid phase. It is respectfully noted that different catalysts are generally used for vapor and liquid phase reactions because the reaction kinetics and catalyst activity are both affected by the pressure and temperature at which the reaction is occurring. Catalyst properties which improve the results of vapor phase reactions cannot, therefore, be presumed to have a similar effect with respect to liquid phase reactions.

If a *prima facie* case of obviousness has been established, which it has not, Applicants have rebutted that case with unexpected results achieved by the use of TEA-mordenite in the liquid phase transalkylation reaction. In this regard, the Examiner's attention is directed to the attached second Declaration by Dr. Wieslaw J. Roth under 37 CFR 1.132, which shows the results of testing comparing a catalyst made in accordance with the present invention and having a particle size of less than 0.5 micron with two commercially available mordenite catalysts also having a particle size of less than 0.5 micron.

As noted in the attached Declaration, Example 1 of Kuchenmeister et al., which employs an unspecified aluminosilicate catalyst having an average crystal size of  $0.41\mu\text{m}$ , the diethylbenzene conversion rate after 1 day on stream is only 11% and that this rate decreases continuously with time on stream until, after 14.6 days, the diethylbenzene conversion rate is 2.8% (an aging rate of about 6%/day). In Comparative Example 1 of Kuchenmeister et al., which again employs an unspecified aluminosilicate catalyst but having an average crystal size of  $0.70\mu\text{m}$ , the diethylbenzene conversion rate after 1 day on stream is even lower, at 4.6%, but this increases or remains generally constant until 6.7 days on stream. The data in Kuchenmeister et al. suggests that the unspecified aluminosilicate catalyst used therein has extremely low activity for diethylbenzene conversion irrespective of crystal size and that, in the case of at least the small crystal material, the catalyst ages rapidly. For both of these reasons, it would be counter-intuitive

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for one of ordinary skill in the art to consider the disclosure in Kuchenmeister et al. in seeking to improve the process of Cheng et al., particularly for commercial application where long run times and hence low aging rates are critical.

Dr. Roth's previous declaration presented data showing that TEA-mordenite having an average crystal size of less than 0.5 micron is more active for the transalkylation of diisopropylbenzene than TEA-mordenite having an average crystal size of greater than 5 micron. These data were batch data generated in an autoclave and did not provide information as to the aging characteristics of the catalysts.

Table 1 of the present Declaration displays the results of a series of tests comparing the aging of the small crystal TEA-mordenite of the present invention with the aging of two commercially available, conventional mordenite catalysts. The tests were run for 7-14 days at a temperature of 260°C, a pressure of 500 psig, a benzene to polyethylated benzene weight ratio of 3:1 and a WHSV of 5. It will be seen from Table 1 that the small crystal TEA-mordenite of the present invention had an initial diethylbenzene (DEB) conversion activity of 77%, significantly higher than that of the Kuchenmeister et al. catalysts, but essentially the same as that of the conventional mordenite catalyst supplied by PQ Corporation. However, although each of the catalysts had a crystal size of below 0.5  $\mu\text{m}$ , the small crystal TEA-mordenite of the present invention exhibited an aging rate significantly lower than either of the conventional mordenite catalysts. This surprising reduction of catalyst aging has significant commercial benefit.

For the foregoing reasons, Applicants respectfully request that the rejection be withdrawn.

#### ***Response to the Rejection of Claim 10***

Claim 10 is rejected under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024, in view of Kuchenmeister et al., EP 733,608, and further in view of Innes et al., U.S. Patent No. 4,891,358.

It is Applicants' position that this rejection fails for the same reasons as given above with respect to the rejection of claims 4 to 9, that there is no *prima facie* case of obviousness because there is no motivation to combine the teachings of Cheng et al. with

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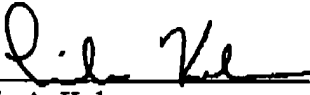
those of Kuchenmeister et al. If a prima facie case of obviousness has been made for the purposes of this rejection, Applicants rely upon the arguments made above with respect to the rejections of claims 4 to 9 and incorporate those reasons herein.

For the foregoing reasons, Applicants respectfully request that this rejection be withdrawn.

### CONCLUSION

Applicants respectfully submit that the foregoing arguments place the application in condition for allowance. Allowance of this application is therefore earnestly solicited.

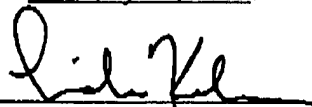
Respectfully submitted,

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I hereby certify that this correspondence is being facsimile transmitted to the United States Patent and Trademark Office, Fax No. (703) 872-9310 on February 14, 2003.

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**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Patent Application of:

W. Roth et al

Serial No. 09/305,019

Filed: May 4, 1999

For: ALKYLAROMATICS PRODUCTION

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**OFFICIAL**

Examiner: Tam Nguyen

Group Art Unit: 1764

**DECLARATION UNDER 37 C.F.R. 1.132**

Commissioner of Patents and Trademarks  
Washington, DC 20231

Sir:

I, the undersigned, Dr. Wieslaw J. ROTH, a citizen of United States, whose address is 123 Boundbrook Ct., Sewell, NJ 0808, do solemnly declare, as follows:

1. I have a Ph. D. in Chemistry (Southern Illinois University, Carbondale, IL; 1981) and 15 years experience in the synthesis and characterization of zeolites and catalysts initially at Mobil Research and Development Corporation and, since the 1999 merger between Mobil and Exxon, at ExxonMobil Research and Engineering Company.

2. I am one of the inventors of the above-identified application and am the same Wieslaw Roth who executed a declaration in connection with this application on October 16, 2002.

3. The present invention, as recited in claim 4, is directed to a process for producing a monoalkylated aromatic compound comprising the steps of:

- (a) contacting an alkylatable aromatic compound with an alkylating agent in the presence of an alkylation catalyst in an alkylation reactor to provide a product

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- comprising said monoalkylated aromatic compound and a polyalkylated aromatic compound, and then
- (b) contacting the polyalkylated aromatic compound from step (a) with said alkylatable aromatic compound in the liquid phase and in the presence of a transalkylation catalyst in a transalkylation reactor separate from said alkylation reactor, said transalkylation catalyst comprising TEA-mordenite having an average crystal size of less than 0.5 micron to produce said monoalkylated aromatic compound. [Emphasis added.]

4. It is my understanding that the Examiner has rejected claims 4 to 9 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and has rejected claim 10 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and further in view of Innes et al., U.S. Patent No. 4,891,458. In particular, I understand that the Examiner relies upon Cheng et al. as disclosing an aromatics alkylation/transalkylation process in which the transalkylation step is conducted in a separate reactor which can contain TEA mordenite. In addition, I understand that the Examiner argues that it would have been obvious to modify the Cheng process by utilizing a TEA mordenite catalyst having an average crystal size of less than 0.5 micron, given that Kuchenmeister et al., page 3, lines 29-30, teach that improved transalkylation control in the production of ethylbenzene is attributable "to the use of an aluminosilicate catalyst having a crystal size less than about 0.50  $\mu\text{m}$  (0.50 microns)." ←

5. I have been asked to comment on the results disclosed in Kuchenmeister et al. and, in particular whether these results would have provided motivation to me, as a skilled researcher in the area of zeolite catalysis, to employ small crystal TEA-mordenite in the process of Cheng et al.



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6. I note that in Example 1 of Kuchenmeister et al., which employs an unspecified aluminosilicate catalyst having an average crystal size of 0.41 $\mu$ m, the diethylbenzene conversion rate after 1 day on stream is only 11% and that this rate decreases continuously with time on stream until, after 14.6 days, the diethylbenzene conversion rate is 2.8% (an aging rate of about 6%/day). In Comparative Example 1 of Kuchenmeister et al., which again employs an unspecified aluminosilicate catalyst but having an average crystal size of 0.70 $\mu$ m, the diethylbenzene conversion rate after 1 day on stream is even lower, at 4.6%, but this increases or remains generally constant until 6.7 days on stream. However, according to Table 2, between 6.7 days and 7.7 days, the conversion rate with the comparative catalyst decreases precipitously to 2.0%. This suggests to me that the data point for 7.7 days with the comparative catalyst may be suspect. More importantly, the data in Kuchenmeister et al. suggests to me that the unspecified aluminosilicate catalyst used therein has extremely low activity for diethylbenzene conversion irrespective of crystal size and that, in the case of at least the small crystal material, the catalyst ages rapidly. For both of these reasons, it would be counter-intuitive to me to consider the disclosure in Kuchenmeister et al. in seeking to improve the process of Cheng et al., particularly for commercial application where long run times and hence low aging rates are critical.

7. In my previous declaration I presented data showing that TEA-mordenite having an average crystal size of less than 0.5 micron is more active for the transalkylation of diisopropylbenzene than TEA-mordenite having an average crystal size of greater than 5 micron. These data were batch data generated in an autoclave and do not provide information as to the aging characteristics of the catalysts. ✓

8. Attached as Table 1 are the results of a series of tests which were conducted by my research team to compare the aging of the small crystal TEA-mordenite of the present invention when used in the transalkylation of a commercial polyethylated benzene feed with the aging of two commercially available, conventional mordenite catalysts. The tests were run for 7-14 days at a temperature of 260°C, a pressure of 500 psig, a benzene

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to polyethylated benzene weight ratio of 3:1 and a WHSV of 5. It will be seen from Table 1 that the small crystal TEA-mordenite of the present invention had an initial diethylbenzene (DEB) conversion activity of 77%, significantly higher than that of the Kuchenmeister et al. catalysts, but essentially the same as that of the conventional mordenite catalyst supplied by PQ Corporation. However, although each of the catalysts had a crystal size of below 0.5  $\mu\text{m}$ , the small crystal TEA-mordenite of the present invention exhibited an aging rate significantly lower than either of the conventional mordenite catalysts.

Table 1

Catalyst	TEA-mordenite	Conventional Mordenite TOSOH HSZ-630HOA	Conventional Mordenite PQ CBV-30A
Silica/alumina ratio	34:1	18:1	51:1
Alpha value	288	84	191
Surface area	427	328	392
Crystal size	<0.5 $\mu\text{m}$	<0.5 $\mu\text{m}$	<0.5 $\mu\text{m}$
Initial DEB conversion	77%	44%	76%
Decrease in DEB conversion/day	0.3%	5.6%	0.9%

9. I consider that the results in Table 1 are unexpected given the teaching in Kuchenmeister et al. and that, in view of the low activity and rapid aging of the Kuchenmeister et al. catalysts, it would not have been obvious to employ a TEA-mordenite catalyst having an average crystal size of less than 0.5 micron in the process of Cheng et al.

The undersigned further declares that all statements made herein of his own knowledge are true and that all statements made on information and belief knowledge

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that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity in the above-captioned application or any patent issuing therefrom.

Wieslaw J. Roth

Dr. Wieslaw J. Roth

2/13/2003

Date